Solution Synthesized Nanostructured Thermoelectric Materials

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Thermoelectric heat engines are currently used in several niche applications for electricity generation and cooling. Many additional applications would be practical if thermoelectric materials with improved figures of merit could be made. Over the past twenty years, many nanostructured materials have been shown to possess improved figures of merit compared to their bulk counterparts mostly due to the reduction in thermal conductivity associated with nanostructured materials. Several classes of solution synthesized nanostructured materials have achieved high figures of merit, yet significant room for improvement exists for solution synthesized nanostructured PbTe.

This thesis describes the solution synthesis of ultra-thin PbTe nanowires at a scale of over ten grams per batch. These materials are washed, dried, and compressed at high temperature to form millimeter-scale nanocomposite discs that, in principle, could be diced into pillars for the traditional thermoelectric device architecture. The properties of unintentionally Te-doped PbTe nanowire-based discs are understood in terms of the phase diagram of the Pb-Te system. The properties are similar to those of bulk PbTe, except for the differences in the mobility and lattice component of thermal conductivity. Unintentionally Te-doped PbTe discs have carrier concentrations that are too low to achieve high figure of merit at high temperature where PbTe is best suited, so two doping strategies using Bi and Na are described, which are shown to effectively raise the carrier concentration to the desired range. Finally, the synthesis of PbTe nanowires is enhanced by replacing the highly toxic reducing reagent, hydrazine, with the relatively benign, ascorbic acid. Nanowires of essentially the same diameter are synthesized using ascorbic acid, albeit at a much slower rate as shown by studies of aliquots taken at different times.

In addition to achieving high figure of merit, it is desirable to create flexible thermoelectric materials as these could be used in a variety of energy harvesting and cooling applications. The recently invented PbTe nanocrystal coated glass fiber composite is one example of a flexible thermoelectric material. Here, the 3ω method is used to determine the thermal conductivity of PbTe nanocrystal coated glass fiber composites in the axial direction. Careful consideration of the effect of radiation helps to ensure more accurate measurement results. Additionally, two of the major issues with PbTe nanocrystal coatings are relieved by replacing PbTe with Ag_2Te nanocrystals. Prototype flexible thermoelectric devices made from Ag_2Te nanocrystal coated nylon mesh are shown to produce over two orders of magnitude more power than similar devices made using PbTe nanocrystal coated glass fibers.